

Effect of oxygen pressure on the orientation of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}/\text{SrTiO}_3$ films deposited on (1 $\bar{1}$ 02) Al_2O_3 substrates

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Abstract

The oxygen pressure P_{O_2} during growth of strontium titanate (SrTiO_3) films on single crystals of (1 $\bar{1}$ 02) oriented sapphire (Al_2O_3) substrates significantly influenced the film orientations. The films were deposited using a pulsed laser (248 nm) deposition process in which the SrTiO_3 films were deposited at a P_{O_2} of either 40 or 200 mTorr, and the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) films were always deposited at 200 mTorr of oxygen. We found that growth at 40 mTorr induced the (110) SrTiO_3 orientation to predominate, while increasing the P_{O_2} to 200 mTorr favored the (100) orientation. YBCO films deposited on these barrier layers were (013) and (001) oriented respectively; these were the orientations that minimized lattice mismatch at the YBCO/ SrTiO_3 interface.

1. Introduction

Replacing normal metal conductors with superconducting films has been shown to improve the performance of microstrip millimeter-wave devices such as filters, resonators and phase shifters [1–4]. The lower dielectric constant of sapphire (Al_2O_3) relative to other commonly used substrates for $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ (YBCO) films (such as LaAlO_3 and Y-ZrO_2) enables devices to be designed so that for a given impedance (e.g. 50 Ω), wider conductor lines can be patterned [5], thus lowering the conductor losses. However, chemical interdiffusion at the YBCO/ Al_2O_3 interface degrades the superconductor, so a barrier layer is usually deposited prior to the superconductor [6]. SrTiO_3 has been shown to effectively limit interdiffusion. To ensure growth of highly (001) oriented YBCO films, it is important to control the orientation of the SrTiO_3 barrier layers. Growth of highly (100) oriented SrTiO_3 films has been reported on (1 $\bar{1}$ 02) Al_2O_3 [7], as well as SrTiO_3 films with mixed (100) and (110) orientations [8]. Because the crystal structures and lattice parameters of SrTiO_3 are very different from those of the (1 $\bar{1}$ 02) Al_2O_3 (SrTiO_3 is cubic with a lattice parameter of 3.90 Å, while $\alpha\text{-Al}_2\text{O}_3$ is hexagonal with $c = 12.99$ Å and $a = 4.76$ Å), a simple one-to-one correspondence of lattice sites and hence epitaxial matching is not possible. It

has been established that many mismatched systems will adopt large surface meshes at the interface in order to maximize the number of coincident lattice sites and thereby minimize the interfacial energy [9, 10]. To what extent lattice matching at the interface and film orientation can be controlled by varying the deposition conditions of various film/substrate combinations has not been established. This paper examines how the oxygen partial pressure P_{O_2} during growth of the SrTiO_3 barrier layer on (1 $\bar{1}$ 02) Al_2O_3 substrates influences the orientation of the SrTiO_3 barrier layer and YBCO films.

2. Experimental procedure

Barrier layer and YBCO films were sequentially deposited at 730 °C using a pulsed layer deposition system. A Questek model 2560 pulsed excimer laser, operating at 248 nm with 30 ns pulses, and 5 pulses s^{-1} was focused on 1-inch diameter SrTiO_3 or YBCO targets with an energy density of 2.5–3.0 J cm^{-2} . The target-to-substrate distance was 4.5 cm. The SrTiO_3 target was fabricated by pressing the SrTiO_3 powders into disks, then firing the disks at 950 °C for 12 h. This target was approximately 65% dense. The YBCO target was obtained from Ceracon, Inc. and was 96% dense. Up to three targets could be mounted on a stainless steel holder. By rotating the target holder, sequential films could be deposited without breaking vacuum or reducing the substrate temperature. This holder did not allow continuous rotation of the target during deposition, but

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the target was moved slightly every 800 pulses to expose a new surface to the laser radiation. The SrTiO_3 and YBCO films were deposited at 730 °C, and the temperature was measured by a thermocouple spot-welded to the heater block. The SrTiO_3 films were 1000 Å thick, and were deposited at either 40 mTorr or 200 mTorr O_2 . The YBCO films were deposited, the chamber was filled with oxygen and the temperature was maintained at 730 °C for 20 min. The films were cooled at 450 °C over 60 min, held at 450 °C for 45 min to ensure complete oxygenation, then cooled to room temperature over the next 45 min.

The films were characterized using d.c. resistance *vs.* temperature, critical current density J_c , scanning electron microscopy (SEM) and X-ray diffraction (XRD) measurements. A Philips model APD 3720 X-ray diffractometer with a Cu source, operating at 40 kV, 20 mA and generating K α radiation of $\lambda = 1.54$ Å was used to determine the orientations of the YBCO and SrTiO_3 films. Peaks within the 2θ range of 5°–65° were detected, and the X-ray detector was rotated at 3 min⁻¹. A JEOL JSM 35C scanning electron microscope, operated at 15–20 kV and typically 100 μA beam current, was used to determine visually the surface morphology and microstructure.

Magnetization J_c values were determined using the Bean model in the cylindrical geometry [11]. The d.c. magnetic fields were applied perpendicular to the film surfaces, and the J_c values were obtained using

$$J_c = \frac{15\Delta M}{r} \quad (1)$$

where J_c is given in A cm^{-2} , ΔM in e.m.u. cm^{-3} and r in cm. The films were patterned as squares, and the radius used in the Bean expression was taken to be half the length of a side.

3. Results

The orientations of the SrTiO_3 barrier layers were extremely sensitive to the P_{O_2} during deposition. XRD (Fig. 1) shows that the SrTiO_3 film deposited at 200 mTorr oxygen was highly (100) textured. The YBCO film was also deposited at 200 mTorr oxygen, and was predominately (001) oriented. Figure 2 shows that SrTiO_3 films deposited at 40 mTorr oxygen were mainly (110) oriented. After the SrTiO_3 deposition, the P_{O_2} was raised to 200 mTorr for the YBCO deposition, and the YBCO film grew with a predominately (013) orientation. Resistance *vs.* temperature measurements (Fig. 3) show that the (001) oriented YBCO film was metallic, with a zero resistance temperature T_0 of 83 K. The normal state resistance of the (013) YBCO films was less metallic, and there was a broad transition to T_0

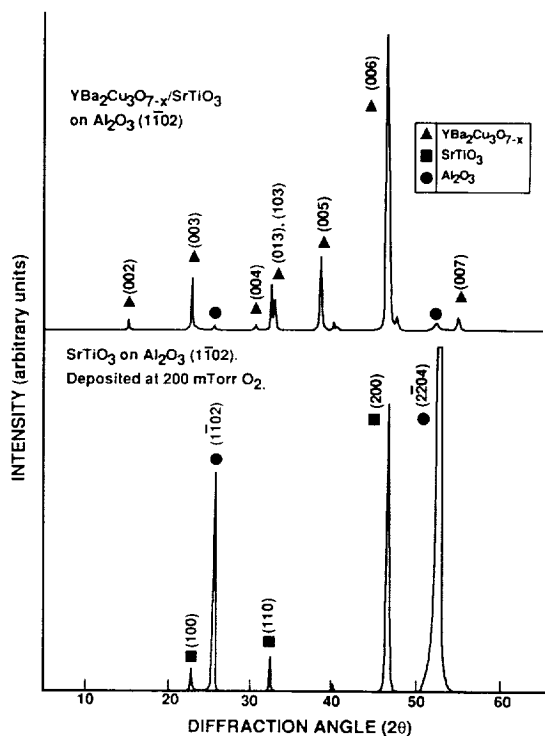


Fig. 1. XRD patterns for SrTiO_3 and YBCO/ SrTiO_3 films deposited on (1102) Al_2O_3 . SrTiO_3 and YBCO films deposited at $P_{\text{O}_2} = 200$ mTorr.

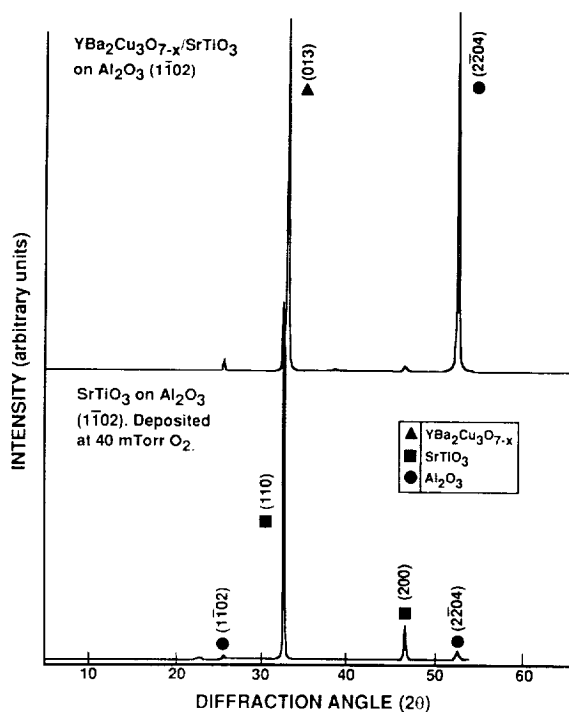


Fig. 2. XRD patterns for SrTiO_3 and YBCO/ SrTiO_3 films deposited on (1102) Al_2O_3 . SrTiO_3 films deposited at $P_{\text{O}_2} = 40$ mTorr, YBCO deposited at $P_{\text{O}_2} = 200$ mTorr.

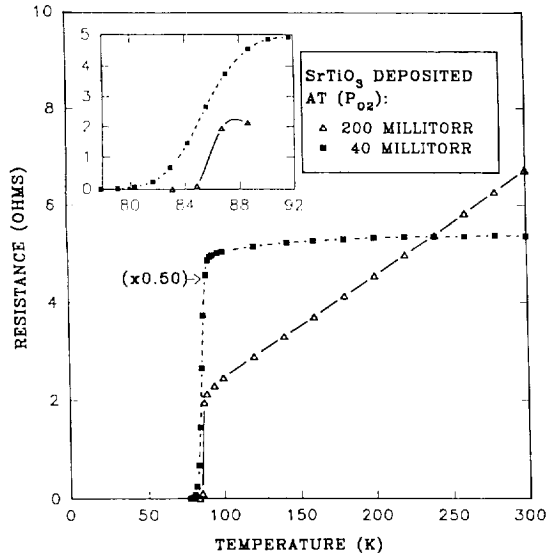


Fig. 3. Resistance vs. temperature data for YBCO/SrTiO₃ films deposited on (1102) Al₂O₃. The orientations of the YBCO films were determined by those of the SrTiO₃, which in turn were determined by the oxygen pressure during the deposition. Resistance values for the (013) oriented YBCO film are multiplied by 0.50.

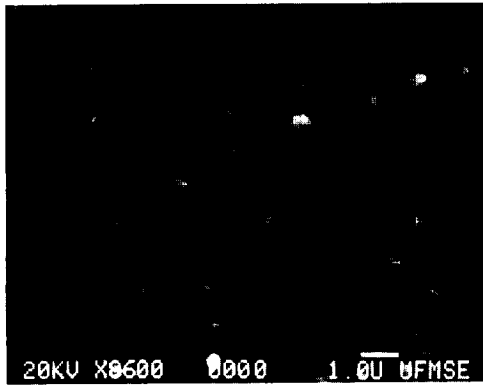


Fig. 4. SEM image of a (001) YBCO/(100) SrTiO₃ film on (1102) Al₂O₃.

at 77 K. The J_c of the (001) YBCO/(100) SrTiO₃ film was $2 \times 10^6 \text{ A cm}^{-2}$, and of the (013) YBCO/(013) SrTiO₃ film was $6.8 \times 10^3 \text{ A cm}^{-2}$ at 4.5 K. The SEM image in Fig. 4 indicates that the (001) YBCO films were highly faceted, but no microcracks were observed. In contrast, Fig. 5 shows that microcracking was prevalent in the (013) YBCO/(110) SrTiO₃ film.

4. Discussion

Figures 1 and 2 show that altering the P_{O_2} during growth of the SrTiO₃ barrier layers had a dramatic influence on the orientations of the films. Normally, we

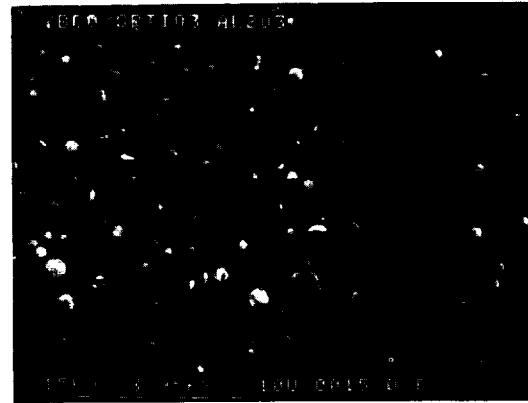


Fig. 5. SEM image of a (013) YBCO/(110) SrTiO₃ film on (1102) Al₂O₃.

expect the film to grow in a manner that minimizes the number of incommensurate lattice sites, so lattice mismatch at the interface is the predominant factor that determines the growth orientation of the film. However, when the lattice mismatch is large, minimization of interfacial energy via lattice matching becomes a less predominant factor. We have calculated that the lattice mismatch of the SrTiO₃/(1102) Al₂O₃ interface along which there is a one-to-one correspondence of lattice sites has a minimum value of 7.7% along the $\langle 110 \rangle$ SrTiO₃// $\langle 1\bar{1}01 \rangle$ Al₂O₃ direction. Perpendicular to this direction, the lattice mismatch along the $\langle 1\bar{1}0 \rangle$ SrTiO₃// $\langle 1\bar{1}20 \rangle$ Al₂O₃ direction is 15.9%. Thus it is reasonable to expect that other deposition parameters heavily influence the film orientation. Henrich [12] showed that oxygen vacancies are the primary defect in SrTiO₃, and minor variations in oxygen content could dramatically alter the Gibbs free energy of formation of the growing film and thus influence the growth orientation.

To see whether this sensitivity to P_{O_2} during deposition was caused only by interactions between the P_{O_2} and SrTiO₃ film or whether the Al₂O₃ substrate also played a role in the growth orientation, SrTiO₃ films were deposited on (100) MgO substrates at an oxygen pressures of 40 mTorr. Other researchers have deposited SrTiO₃ film on MgO substrates at oxygen pressures to 200 mTorr [13]. In both cases, the SrTiO₃ films were (100) oriented, indicating that matching of the lattice types and constants played a more dominant role in determining the film orientation than in the case of SrTiO₃ films on (1102) Al₂O₃ substrates.

Minimization of the lattice mismatch at the YBCO/SrTiO₃ interface dominated the orientations of YBCO films deposited on the SrTiO₃ barrier layers. The temperature dependence of the resistance of the (001) YBCO film was similar to that of the films on (100) SrTiO₃ substrates; they had good normal state properties but a slightly depressed T_0 . However, the normal state resistance and T_0 for the (013) film were inferior

to those reported for (013) films deposited on (110) SrTiO_3 substrates [14]. This was attributed to microcracking in the YBCO film. Since the thermal expansion coefficient of YBCO is highest in the $\langle 001 \rangle$ direction [15], thermally induced stresses were higher in the (013)-oriented than the (001)-oriented YBCO film, and thus more prone to microcracking.

5. Conclusions

We have shown that altering the P_{O_2} during deposition of the SrTiO_3 films on (1102) Al_2O_3 dramatically changed the growth orientations of the films. Although the mechanisms that dictated the film's orientations are difficult to determine, it was established that the growth orientation was a result of synergistic interactions between the film, substrate and P_{O_2} during deposition.

We believe the sensitivity to P_{O_2} resulted from the lack of epitaxy along low-index directions, and thus minimization of the interfacial energy via lattice matching was not the dominant parameter that determined the growth orientations. Other factors such as symmetry between the SrTiO_3 and Al_2O_3 crystal structures may also be partially responsible for the sensitivity of film orientation to oxygen pressure.

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